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EDDY DAMPING BY SURFACE ACTIVE AGENTS IN INTERFACIAL TURBULENCE

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Abstract—In the present work, a study has been made of the nature of interfacial resistance to mass transfer caused by films of surface active substances at the liquid–liquid interface or liquid–gas interface in an agitated cell, since at a clean interface there is conclusive evidence that no resistance of any industrial consequence arises, but transport phenomena is markedly damped across interfaces with such films. The paper purports that the frequency damping of eddy penetration into the turbulent interface is not the decisive factor in the retardation of transport phenomena across film covered interfaces, but the area of clearance of film molecules by the penetrating eddy at the interface. The eddy is modeled as a lump of fluid which is impelled into the interface and disturbs the film's molecules over a "Prandtle mixing length" which retaliates by means of its elasticity, thereby resisting the clearance of each eddy. On this basis, a formula has been obtained for the amount of clearance of area, hence the amount of area renewal, and the damping of transport phenomena which incorporates the surface compressional modulus of elasticity.

INTRODUCTION

In recent years interfacial turbulence has assumed paramount importance in engineering, since there is direct impact on mass, momentum, and heat transfer at turbulent interfaces. In order to study the effect of eddies on engineering problems, stirred cells are usually employed, and for this reason, the structure of turbulence in a stirred cell, and, in particular, at the interface, is the domain of interest. Although surface renewal theories have gained general acceptance for transport phenomena across turbulent interfaces and the damping of eddies at such interfaces is definitely established as hydrodynamic in nature, the mechanism by which the hydrodynamic damping is effective is still in doubt and the effect of the surface compressional modulus of elasticity due to a spread film of surface active agents has been very elusive. Most of the existing theories are concerned with eddy penetration into the interface and the effect of surface conditions on the hydrodynamics of the system.

In the present work, a study has been made of the nature of interfacial resistances to mass transfer caused by films of surface active substances at the liquid–liquid interface, or liquid–gas interface in an agitated cell. In recent years, attention has been drawn to the possibility of such interfacial resistances, and it seems fairly conclusive now, that, for a clean interface there are no resistances of any industrial consequence, but in the presence of surface active materials there is a marked retarding effect on mass transfer.

Experiments with talc covered interfaces support the surface renewal theories, since eddies have been observed to clear local areas of the interface. Further, these observations

point to the acceptance of hydrodynamic damping of surface turbulence for the nature of interfacial resistances.

We conclude that surface renewal mechanism is valid in such systems, and, that interfacial resistances must necessarily be of a hydrodynamic nature, the eddy disturbances being damped by the presence of spread films or insoluble films.

On this basis, it is possible to formulate an expression for the total area of eddy exposure, which is synonymous with the total area of surface renewed and which incorporates the action of the film directly on the hydrodynamics of the problem of virtue of its retarding effect on the tangential surface motion through its compressional modulus of elasticity C_s^{-1}.

BOUNDARY CONSIDERATIONS

Under given conditions, tangential forces may be exerted on the surface of a liquid, along with the normal pressure. If the surface tension changes from point to point, a tangential force will be exerted, which is determined by the surface tension gradient. The tangential force per unit area

$$P_t = \text{grad } \gamma, \tag{1}$$

the positive sign indicating that the force P_t tends to move the surface from smaller to higher surface tension, or from greater to lower concentration of film molecules.

At an interface which is contaminated the boundary conditions are unaltered except for the continuity of the shear stresses. We find that

$$T + P_t = T^1, \tag{2}$$

where T and T^1 are the tangential viscous stresses for the two phases forming the interface.

It follows that in the case of a static liquid system

$$T^1 = T = 0, \tag{3}$$

and

$$P_t \neq 0, \tag{4}$$

hence these boundary conditions cannot be satisfied. We conclude that surface tension gradients must necessarily set the liquid in motion. In the case of a constant surface tension where the molecules of the film are evenly distributed, disturbances from the bulk liquid would create surface tension gradients, hence surface forces, which would then resist any tangential motion of the disturbance through the surface compressional modulus C_s^{-1} defined by

$$C_s^{-1} = A \frac{d\gamma}{dA}. \tag{5}$$

In the damping of waves by films, Dorrestein[6] considered the shear stress

$$T = P_t = \frac{dT_{xx}}{dx}, \tag{6}$$

where T_{xx} is the stress tensor at the surface analagous to the stress tensors in the bulk of fluid motions.

Therefore,

$$T_{xx} = \gamma + \mu_s \frac{\delta u_s}{\delta x}, \tag{7}$$

where γ is the surface tension and μ_s the surface viscosity, u_s is the velocity along the surface. Dorrestein[6] therefore, made allowance for the dissipation of energy due to surface viscosity by incorporating this term into the surface stress tensor T_{xx}. Mayers[14] however, discovered that mass transfer was appreciably damped before surface viscosity was measurable. If the mass transfer damping is a consequence of hydrodynamic damping brought about by spread films, then the surface compressional modulus of elasticity must be the important controlling factor. Following Mayers[14] we can neglect the surface viscosity term and write the stress tensor

$$T_{xx} = \gamma, \tag{8}$$

and

$$T = P_t = \frac{d\gamma}{dx}. \tag{9}$$

Dorrestein[6] derived the equation

$$\frac{1}{A}\frac{dA}{dt} = \frac{1}{C_s^{-1}}\frac{d\gamma}{dt} = \frac{\delta u_s}{\delta x} = \mathrm{div}(u_s), \tag{10}$$

in which u_s the velocity along the surface gave the only contribution to the change in area assuming that the film molecules adhered firmly to the underlying liquid.

The effect of surface active substances on liquid flow is apparent as long as the equation

$$T + P_t = T', \tag{11}$$

is controlling. This condition maintains consistency between the viscous stresses and the surface forces created by the film. At very high Reynolds numbers when large scale motions are predominant and viscosity of no consequence, surface films would be ineffective in damping the motion, as they would be swept away. In this light, the assumption of viscous eddy motion at the interface would gain support since Mayers[14] observed damping in mass transfer rates between the range of Reynolds number between 1000 and 4500. If, on the other hand, large-scale motions are propagated into the interface, the surface film would be swept away and damping, if any, would be insignificant. Since mass transfer is now visualised as necessarily involving surface renewal, and since the effect of the film is visualised as one of hydrodynamic impediment, the assumption of viscous eddy penetration into the interface can be made very plausible indeed.

Let us consider the mass balance equation for the film molecules whenever an eddy disturbance occurs. The distribution of film molecules is affected by several factors. First of all, the surface-active molecules are entrained by liquid flow, assuming no slip the molecules of the film would adhere firmly to the underlying liquid and dragged along with the ensuing motion. Consequently, a convective flux of surface active substance arises

along the liquid surface giving

$$j_{\text{surf}} = N_s V, \tag{12}$$

where V is the tangential liquid velocity at the surface of the disturbance.

If the concentration of surface active material varies from point to point along the surface, a diffusion of film molecules would follow, and proceeds from points of higher to points of lower concentration. This diffusional flux is measured by

$$j_{\text{diff}} = -D_s \text{ grad } N_s, \tag{13}$$

where D_s is the surface diffusion coefficient.

If the surface-active material is soluble in the liquid phase, it can pass from the surface phase into the bulk phase, or vice-versa, whereby a mass flux designated by j_n would arise.

The law of conservation of surface-active material, in general, can now be written as

$$\frac{\delta N_s}{\delta t} + \text{div } (j_{\text{surf}} + j_{\text{diff}}) + j_n = 0. \tag{14}$$

In this particular problem, the film is insoluble, so that

$$j_n = 0. \tag{15}$$

The distribution of film molecules is uniform until the film is provoked locally by eddy disturbances. We may, therefore, neglect the diffusional flux especially as the film molecules are envisaged as sticking to the underlying liquid and moving with it.

With the above considerations in mind the mass balance equation becomes

$$\frac{\delta N_s}{\delta t} = - \text{div } (j_{\text{surf}}), \tag{16}$$

that is,

$$\frac{\delta N_s}{\delta t} = - \text{div } (N_s V), \tag{17}$$

which can be written as

$$\frac{dN_s}{dt} + N_s \text{ div } (V) = 0, \tag{18}$$

For slow motion eqn (18) can be written as

$$\frac{\delta N_s}{\delta t} + N_s \text{ div } (V) = 0, \tag{19}$$

analagous to the equation of continuity in fluid flow given by

$$\frac{d\rho}{dt} + \rho \text{ div } (V) = 0. \tag{20}$$

The surface tension γ is a function of the surface concentration of film molecules N_s, therefore,

$$\gamma = \gamma (N_s), \tag{21}$$

so that

$$\frac{d\gamma}{dt} = \frac{d\gamma}{dN_s} \frac{dN_s}{dt}, \tag{22}$$

Substituting for dN_s/dt from above, we get

$$\frac{d\gamma}{dt} = - \frac{d\gamma}{dN_s} N_s \text{ div } (V_t). \tag{23}$$

By definition,

$$C_s^{-1} = A \frac{d\gamma}{dA}, \tag{24}$$

where A is the area available per molecule, therefore,

$$N_s = 1/A. \tag{25}$$

Hence,

$$C_s^{-1} = A \frac{d\gamma}{dN_s}, \tag{26}$$

and

$$\frac{d\gamma}{dt} = C_s^{-1} \text{ div } (V). \tag{27}$$

This is the same equation obtained by Dorrestein[14] following the motion of the surface. This implies that the Dorrestein equation is, in effect, a mass balance equation for the film molecules.

Levich[12] has also shown that the surface compressional modulus of elasticity C_s^{-1} can be defined by $N_s \mid d\gamma/dN_s \mid$.

FORMULATION OF THE PROBLEM

Let us assume for convenience that the viscous eddies penetrating into the interface are spherical bodies of fluid. This assumption is not completely without foundation, since the observed clearances of the talc particles conform to approximately circular shapes, suggesting that the body of fluid causing a local disturbance possesses spherical symmetry.

Since we are neglecting the complicated effects experienced by the viscous eddy on entry into the interface (such as distortion and uneven drags over its surface) we can assume that the motion into the interface takes place smoothly. Now, during the flight of the viscous eddy into the interface from the bulk phase no definite boundary can exist around the eddy since surface tension is non-existent for a volume of fluid moving in the

same medium. However, our domain of interest is at the interface itself, hence, we can disregard any previous history of the viscous eddy and consider its effect in clearing the interface locally.

The problem would now appear to be one to which Newton's second law (which states that

$$p = mf, \tag{28}$$

where p is the applied force and f the acceleration) is applicable. For a contaminated interface the tangential motion around the disturbed area will be opposed by the elastic property of the film, but for a clean interface this tangential motion would occur without inhibition, since any dissipation of energy by viscosity is neglected. However, this approach is not possible directly, because we are unable to introduce mass into the problem, but an analysis based on the consideration of shear stress from the eddy of motion just beneath the surface provides a formula resembling the equation of Newton's second law without having recourse to the mass of a particle of eddy fluid moving with the disturbed film molecules.

Whenever an eddy enters the interface giving rise to a small elevation it must induce some motion in the upper phase regardless of whether the interface is liquid–liquid or liquid–gas. Levich[12] has shown that the motion of small gas bubbles in a liquid medium obey the Rybczyn'ski-Hadamard[7] formula. This implies that even for a gas–liquid interface the continuity of shear stress must be preserved across the interface. However, the gas phase can only sustain small shear stresses so that the continuity of shear stress across this interface implies further that motions are small, or that the Reynolds number is low, being less than or equal to unity.

In the most general case when surface active agents are spread on the surface, the shear stress from the eddy of motion beneath the film would be utilized in two ways. Firstly, to overcome the elastic property of the film molecules in clearing the locally disturbed area, and then to induce a motion in the upper phase. The original motion from beneath the interface is viscous and therefore small; the induced motion is therefore even smaller and so negligible in the general case. The small viscous shear stress from the eddy of motion beneath the surface is therefore used primarily in clearing the film molecules, but does not vanish in the absence of surface active agents.

Following Dorrestein[14], we can write

$$T = P_t + T^1, \tag{29}$$

where T is the shear stress exerted on the surface from the liquid beneath, T^1 the shear stress due to the motion induced above and P_t the tangential force created by the film molecules under compression. For a clean surface

$$P_t = 0, \tag{30}$$

therefore,

$$T = T^1. \tag{31}$$

And for the general case above

$$T = P_t, \tag{32}$$

since T^1 due to the induced motion in the upper phase is negligible. Again, following Dorrestein[14]

$$P_t = \text{grad } \gamma, \tag{33}$$

hence

$$T = \text{grad } \gamma. \tag{34}$$

The mass balance equation for the film molecules has been derived in the previous section in the form

$$\frac{d\gamma}{dt} = C_s^{-1} \text{ div } (V), \tag{35}$$

where V is the tangential velocity at the disturbed area around the eddy.

If the eddies are propagated by virtue of the fluctuating corrugated boundaries of the turbulence, whereby its viscous interface protrudes into the surface, then the life-times of these disturbances are controlled by only the contractions and expansions within the turbulent core, and from its statistical nature the average life-times of these disturbances are constant, and completely independent of surface conditions. During the time spent by the disturbance in the interface the tangential motion around the portion of eddy above the interface would drag the film molecules, thereby clearing the talc covered interface locally. However, the compressional modulus of elasticity would then become operative and resist the tangential motion according to the law

$$V = V_0 \, e^{-C_s^{-1} t/\mu\lambda}, \tag{36}$$

for an eddy, as a viscous drop with a hypothetical velocity distribution.

Since the elevation caused by the eddy is small we may write

$$V \doteq \frac{dx}{dt}, \tag{37}$$

where x is measured along the surface of the eddy. Hence

$$\frac{dx}{dt} \doteq V_0 \, e^{-C_s^{-1} t/\mu\lambda}. \tag{38}$$

Integration with the initial condition

$$x = 0, \quad \text{at } t = 0, \tag{39}$$

yields

$$x \doteq \frac{V_0 \mu\lambda}{C_s^{-1}} [1 - e^{-C_s^{-1} t/\mu\lambda}]. \tag{40}$$

This represents the clearance along a line in the surface, but the body of fluid is spherical,

hence the area cleared in the surface can be measured by

$$x^2 \doteq \left[\frac{V_0\mu\lambda}{C_s^{-1}}(1 - e^{-C_s^{-1}t/\mu\lambda})\right]^2 . \tag{41}$$

Let us examine

$$x \doteq \frac{V_0\mu\lambda}{C_s^{-1}}[1 - e^{-C_s^{-1}t/\mu\lambda}], \tag{42}$$

at low and high values of C_s^{-1}. We can write the exponential function as a series giving

$$x \doteq \frac{V_0\mu\lambda}{C_s^{-1}}\left[\frac{C_s^{-1}t}{\mu\lambda} + \left(\frac{C_s^{-1}t}{\mu\lambda}\right)^2 \pm \text{higher powers of } C_s^{-1}\right] . \tag{43}$$

Therefore,

$$x \doteq V_0\left[t + \frac{C_s^{-1}}{\mu\lambda}t^2 \pm \text{higher powers of } C_s^{-1}\right] , \tag{44}$$

which gives

$$x \doteq V_0 t, \tag{45}$$

for

$$C_s^{-1} = 0, \tag{46}$$

and $x \to 0$ for $C_s^{-1} \to \infty$ so that the clean surface becomes a special case of the general theory as $C_s^{-1} \to 0$.

Let us examine further the function

$$x = \frac{1}{C_s^{-1}}(1 - e^{-C_s^{-1}}), \tag{47}$$

to determine the turning points if there exist any. The required condition for turning points is given by

$$\frac{dx}{dC_s^{-1}} = 0 = \left(\frac{-1}{C_s^{-1}}\right)^2(1 - e^{-C_s^{-1}}) + \frac{e^{-C_s^{-1}}}{C_s^{-1}} \tag{48}$$

Hence,

$$C_s^{-1}\, e^{-C_s^{-1}} = 1 - e^{-C_s^{-1}} \tag{49}$$

or,

$$e^{C_s^{-1}} = 1 + C_s^{-1}. \tag{50}$$

Therefore,

$$1 + C_s^{-1} + (C_s^{-1})^2 + (C_s^{-1})^3 + \cdots = 1 + C_s^{-1}, \tag{51}$$

so that

$$(C_s^{-1})^2 + (C_s^{-1})^3 + (C_s^{-1})^4 + \cdots = 0 \tag{52}$$

Since C_s^{-1} can only assume values from 0 to ∞

$$C_s^{-1} = 0 \tag{53}$$

provides the only turning point in the function

$$x = \frac{1}{C_s^{-1}}(1 - e^{-C_s^{-1}}). \tag{54}$$

This turning point must obviously be a maximum since the function is plotted over the range C_s^{-1} from 0 to ∞ and tends to zero at $C_s^{-1} \rightarrow \infty$. It is now evident that the amount of clearance of each eddy disturbance is reduced with the increase in C_s^{-1} at a constant rate of stirring, since all other quantities in x^2 are constants.

On this basis, it is possible to formulate an expression for the total area of eddy exposure, which is synonymous with the total area of surface renewed and which incorporates the action of the film directly on the hydrodynamics of the problem by virtue of its retarding effect on the tangential surface motion through its compressional modulus of elasticity C_s^{-1}.

The theory, in essence, considers an eddy placed at the interface without regard for its previous history. Also, its ability to clear a local area of surface by means of its tangential velocity opposed by the elastic property of the film is examined. Eddies can be propagated into the interface in any manner, especially if its mode of propagation is due to the fluctuating corrugated boundaries of the turbulent volume of liquid, but in all cases the shear stress on the interface is of the order of magnitude $\mu V/\lambda$, so that the tangential damping of the spread film remains the same.

We may extend the above analysis to include all kinds of eddies, viscous or non-viscous, which is the crux of the problem. The boundary condition is

$$\frac{dT}{dt} = \text{grad}\,(C_s^{-1}\ \text{div.}\ V), \tag{55}$$

where V is the velocity of an eddy along the surface since the velocity perpendicular to the surface is negligibly small, because the hydrodynamic damping is effected by the surface compressional modulus of elasticity C_s^{-1}, which acts along the surface and renders the small vertical components of velocity ineffective. Using this idea of the kind of boundary condition for viscous eddies, the shear stress T over a prandtl mixing length is of the form $\mu V/\lambda$ where μ is the viscosity of the liquid and λ the size of the eddy. Therefore, $d/dt\ (\mu V/\lambda)$, which is the rate of change of shear stress over a prandtl mixing length is opposed by the resistive effect of the compressional modulus through grad $(C_s^{-1}\ \text{div.}\ V)$ which must be of the form $C_s^{-1}\ V/\lambda^2$ over a prandtl mixing length for any kind of eddy and is dimensionally consistent.

Hence,

$$\frac{d}{dt}\left(\frac{\mu V}{\lambda}\right) = -\frac{C_s^{-1} V}{\lambda^2}, \tag{56}$$

and therefore

$$V = V_0\, e^{-C_s^{-1} t/\mu\lambda}, \tag{57}$$

Hence the damping of the cleared area, which is synonymous with the damping of the area renewed.

APPLICATION TO MASS TRANSFER

In order to apply the theory of mass transfer it is necessary to calculate the total area renewed.

In general, the mean rate of absorption per unit area is given by

$$R' = \int_0^\infty \phi(t)\,\psi(t)\,dt = \int_0^\infty \phi(t)\,dQ(t), \tag{58}$$

where $Q(t)$ is the amount of absorption per unit area in time t.

In this particular case when the agitation is constant and, eddies are visualised as having the same average strengths and life-time, then

$$\phi(t) = f(\Theta),\ 0 < t < \Theta, \tag{59}$$

and

$$\phi(t) = 0, \tag{60}$$

for $t > \Theta$.

This is a step function as in the Higbie's[8] case and is constant in time, hence

$$R' = \phi(t) \int_0^\infty \psi(t)\,dt = \phi(t)\,Q(t), \tag{61}$$

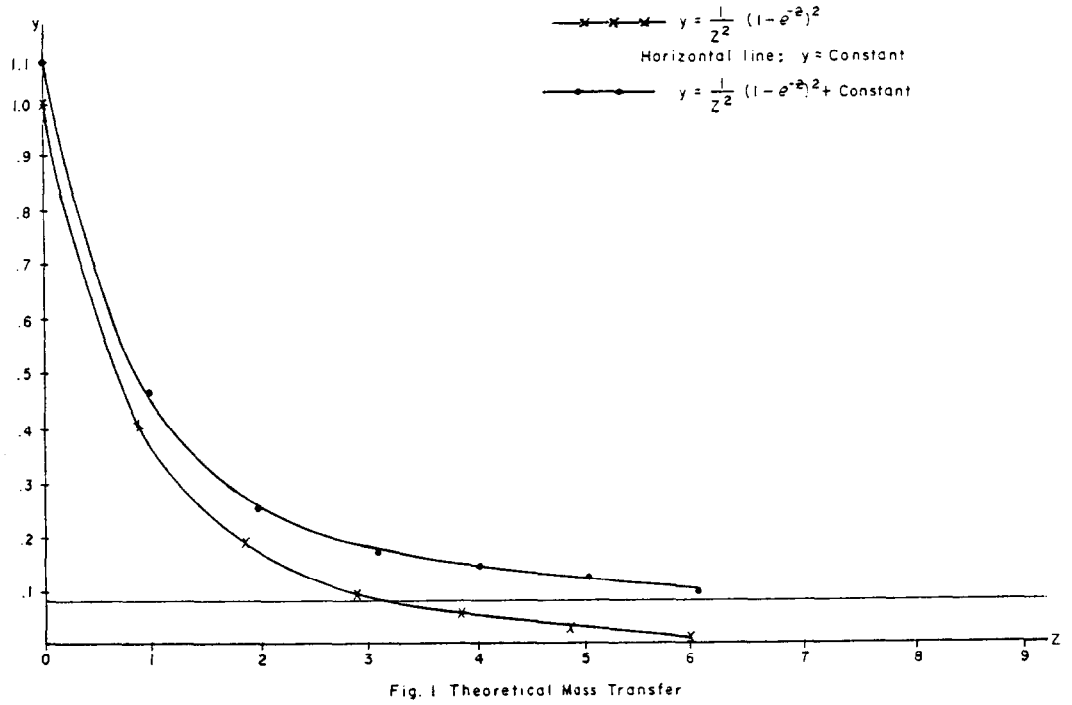

Fig. 1. Mass transfer curves. (The line -X-X-X- represents the theoretical mass transfer curve $y = 1/z^2\,(1 - e^{-z})^2$ which represents the damping in surface renewal, hence mass transfer of the form $MD^{1/2}\,f(C_s^{-1})$. The horizontal line represents mass transfer over the stagnant portions of the form HD^1. The line -O-O-O- represents the resultant mass transfer curve of the form $K = MD^{1/2}\,f(C_s^{-1}) + HD^1$ since both processes are simultaneously operative.

where R' is the average rate of absorption. Now

$$\phi(t) = f(\Theta) \propto n \cdot f_s x^2, \tag{62}$$

and $Q(t)$ is proportional to $\sqrt{Dt}/\pi$ as in the Danckwerts[2] theory, where n is the number of eddies contained in the spectrum that penetrates into the interface, f_s the frequency of the penetrating spectrum and x^2 the area of exposure of each eddy. Therefore, the average rate of absorption over the total area renewed becomes

$$R' \propto n \cdot f_s x^2 \frac{\sqrt{Dt}}{\pi} \tag{63}$$

that is,

$$R' \propto n \cdot f_s \frac{\sqrt{Dt}}{\pi} \left(\frac{\mu\lambda V_0}{C_s^{-1}}\right)^2 (1 - e^{-C_s^{-1}t/\mu\lambda})^2. \tag{64}$$

At a constant rate of agitation, then, n, f_s, λ, V_0 and t are all constants. Hence the average rate of absorption R' changes according to the above law with the corresponding variation in C_s^{-1}.

In general then

$$R' \propto f(Re)\, g\,(C_s^{-1}) D^{1/2}, \tag{65}$$

where Re = Reynolds number. The mass transfer for the surface renewal part is a transient one depending on $D^{1/2}$ for all surface conditions, and would follow the pattern of the curve drawn in Fig. 1 at a constant stirring speed. At the same time, there will always be mass transfer taking place over the portions of surface that are not renewed. If we consider that over these stagnant portions mass transfer takes place steadily, then this contribution would be of the form HD^1, giving a dependence of D^1 at all times.

The total mass transfer taking place is then given by $MD^{1/2} f(C_s^{-1}) + HD^1$, where M and H are arbitrary constants at a constant rate of agitation, since these two processes are simultaneously operative. The resultant effect is also demonstrated on Fig. 1. This holds for all surface conditions and tends to $MD^{1/2} + HD^1$ for a clean surface being a special case of the general theory since $f(C_s^{-1}) \to 1$, as $C_s^{-1} \to 0$ and $f(C_s^{-1}) \to 0$ as $C_s^{-1} \to \infty$, so that mass transfer approaches the stagnant layer theory asymptotically. This implies that surface renewal as formulated here becomes the stagnant layer theory in the limiting case of surface active material where the compressional modulus of elasticity C_s^{-1} becomes infinite in value. Experimentally, however, one would measure a constant amount of mass transfer at some finite value of C_s^{-1} onwards, since small differences are not measurable.

CONCLUSIONS

Resistances seem to occur whenever a film is spread over a clean surface, this additive resistance decreases mass transfer from that of a clean surface down to that of a stagnant layer with increasing film strength.

The present theory is consistant with experimental observations, in that, it allows for eddy penetration into the interface. It considers that interfacial resistances are brought

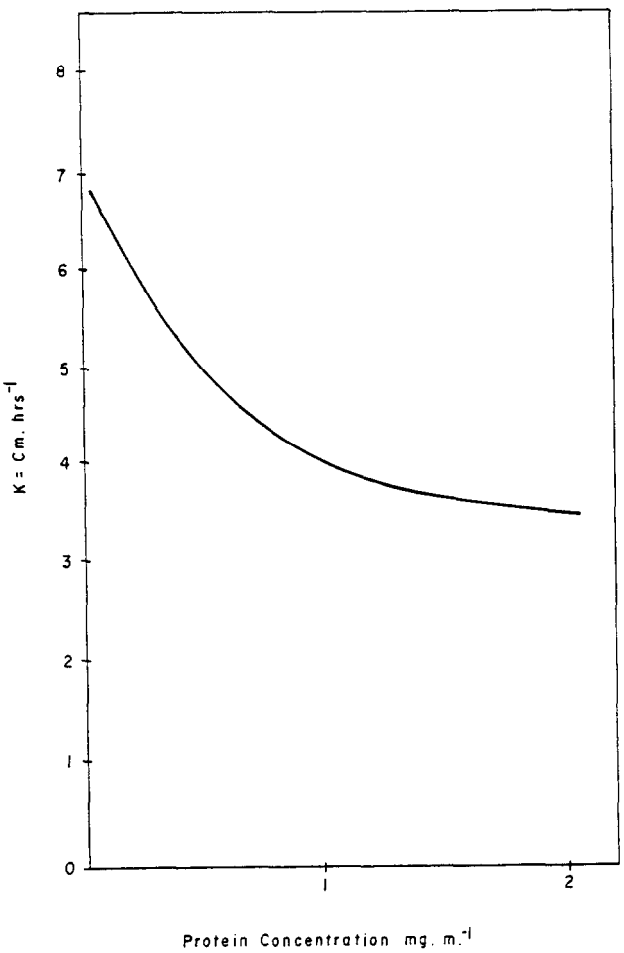

Fig. 2. Mass transfer damping with protein concentration (Kilner[9]).

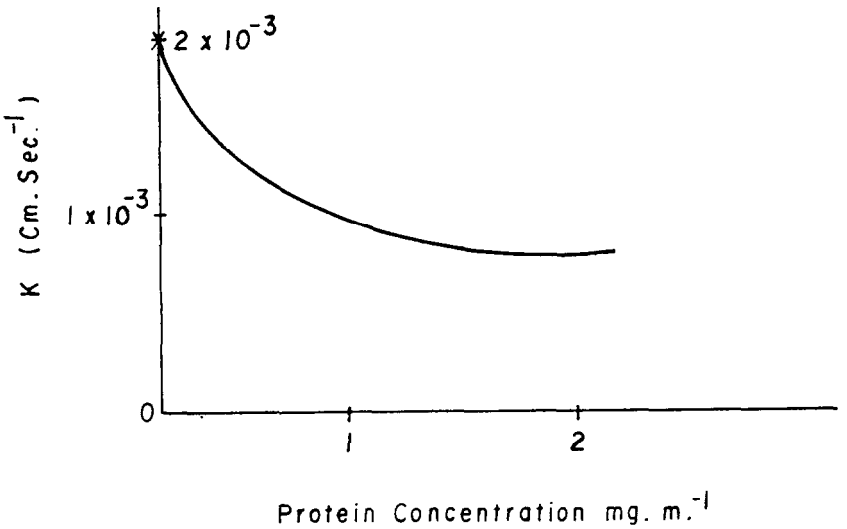

Fig. 3. Mass transfer damping with protein concentration (Mayers[13]).

about by hydrodynamic interference through the tangential damping of the eddy motion by the surface compressional modulus C_s^{-1}.

The new model of an eddy exposure area incorporates the effect of C_s^{-1} directly on the tangential hydrodynamic damping of the eddy motion for the first time and the predicted theoretical mass transfer curve Fig. 1 compares favourably with the experimental mass transfer curves of Kilner and Mayers, Figs. 2 and 3.

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